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PUBLIC HEALTH ASPECTS OF INCREASING TETREATHYL LEAD CONTENT IN MOTOR FUEL

Public Health Service
Washington, D.C.

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PUBLIC HEALTH ASPECTS OF INCREASING TETRAETHYL LEAD CONTENT IN MOTOR FUEL

A REPORT
Advisory Committee on
Tetraethyl Lead to
Surgeon General of
Public Health Service

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Springfield VA 22151

U.S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
PUBLIC HEALTH SERVICE
Bureau of State Services
Division of Special Health Services
Occupational Health Program

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Preface

In response to the expressed interest of the petroleum industry in increasing the tetraethyl lead content in motor fuel, an advisory committee was convened by the Surgeon General of the Public Health Service on January 8 and 9, 1959. The findings of this expert group are presented in this report.

Since the Public Health Service has no legal authority to control the concentration of lead in gasoline, the committee's findings should not be interpreted as a regulation of the Public Health Service or as an agreement with the industry.

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Committee's Letter of Recommendation

MARCH 30, 1959.

SURGEON GENERAL LEROY E. BURNEY,
United States Public Health Service,
Washington, D.C.

SIR: The Committee appointed at your request to consider the public health hazard resulting from the proposed increase of tetraethyl lead from a maximum of 3 to a maximum of 4 cc. per gallon of motor fuel met in Washington on January 8 and 9, 1959. Dr. H. J. Magnuson, Chief, Occupational Health Program, Public Health Service, served as chairman, and Mr. Henry N. Doyle, Assistant Chief, acted as reporter.

Prior to the meeting, the Committee members reviewed Public Health Bulletin 158, "Proceedings of a Conference to Determine Whether or Not There Is a Public Health Question in the Manufacture, Distribution or Use of Tetraethyl Lead Gasoline" (1925), Bulletin 163 "The Use of Tetraethyl Lead Gasoline in its Relations to Public Health" (1926), and other related scientific publications made available by the Occupational Health Program. Additional background information was provided by one of the Committee members, Dr. William P. Yant, who had participated in the 1925 and 1926 conferences called by the Public Health Service to consider the health problem related to tetraethyl lead. These two previous conferences had also been attended by Dr. Robert A. Kehoe, who participated in the first day of the present meeting as medical consultant to the Ethyl Corp.

On January 8, the Public Health Service representatives defined the scope of the Committee's responsibilities and reviewed the history of the tetraethyl lead problem and the relationship of the Public Health Service to the manufacturers and distributors of tetraethyl lead.

Representatives of the Ethyl Corp. and the E. I. duPont de Nemours & Co. presented technical and medical data relating to the proposed increase. The Ethyl Corp. estimated that should the recommended increase be effected, the concentration of tetraethyl lead in gasoline by 1963 would be 0.4 cc. per gallon greater than would be the concentration if the 3-cc. limit were continued. Without the increase, it was estimated that the tetraethyl lead in the U.S. pool gaso-

line would rise from 2.09 cc. to 2.70 cc., and with the increase, to 3.07 cc. per gallon. Estimates by the duPont Co. were slightly lower than those offered by the Ethyl Corp.

On January 9, the Committee met in executive session with technical representatives of the Public Health Service. The proceedings of the meetings and a summary of the data presented to the Committee are contained in the attached report.

The Committee wishes to point out that a conclusive answer is impossible at the present time because of the lack of medical data. While the data available to the present Committee were far more complete than those on which the 1926 Committee had to make its decision, the present Committee was concerned about the inadequacy of medical data derived from human exposures on which to base a conclusive answer to the problem. It is regrettable that the investigations recommended by the Surgeon General's Committee in 1926 were not carried out by the Public Health Service. If data were now available on body lead burdens, with 1926 as a baseline, a more objective decision would have been possible. Such studies should be undertaken without further delay to assure the validity of the present decision and to guide future committees, particularly since some authorities believe that the tolerable limit of lead absorption is being approached.

The Committee recognized that the permissive amount of lead in drinking water may be lowered in the near future. Such action can be taken in view of the improvement in treatment methods for the removal of lead from water. The Committee would welcome similar efforts to reduce the amounts of lead in the atmosphere and was particularly interested in the reported research on the filtration of lead compounds from automobile exhaust in connection with studies of catalytic oxidizers.

The Committee concluded that (1) a change in the maximum concentration of tetraethyl lead in motor fuel from 3.0 to 4.0 cc. would not increase the hazards involved in the manufacture and distribution of leaded gasoline and (2) available data do not indicate that such change would significantly increase the hazard to public health from air pollution.

Should the industry increase the concentration of TEL in gasoline, it is the recommendation of your Committee that:

1. The industry proceed cautiously with the increase so that the increase in any one year in the tetraethyl lead content of U.S. pool gasoline not exceed 0.25 cc., and the concentration of tetraethyl lead in U.S. pool gasoline not be more than 3.2 cc. per gallon at the end of five years.

2. The industry supply annually to the Public Health Service the domestic consumption figures for tetraethyl lead.

3. The Public Health Service, in collaboration with the manufacturers of tetraethyl lead, the petroleum industry, and other appropriate organizations, conduct studies to provide more definitive data on levels and trends of atmospheric lead contamination in selected urban areas and of the body burden of lead of selected population groups. If these studies should bring to light new data of significance, you may wish to reconsider the recommendation of this Committee.

Respectfully,

LEONARD GREENBURG, M.D. W. P. YANT, D. Sc.
H. H. SCHRENK, Ph. D. S. H. OSBORN, M.D.
J. H. STERNER, M.D.

HENRY N. DOYLE, *Recorder*
HAROLD J. MAGNUSON, M.D., *Chairman*

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Introduction

Early in 1923 tetraethyl lead (TEL) began to be used commercially as a gasoline additive to increase the efficiency of internal combustion motors. The apprehension of public health officials over such wide distribution of a toxic lead compound was intensified when a number of fatal poisonings occurred in the manufacture and mixing of TEL. At the suggestion of the Surgeon General, the distribution of TEL and the sale of leaded gasoline were temporarily discontinued on May 5, 1925.

On May 20, 1925, the Surgeon General called a conference of public health and interested authorities for "the determination of such facts and the reporting of such investigations and experiences as may lead to a definite determination as to the hazards, if any, which follow the manufacture, distribution, and use of TEL and similar substances, not only to those who are engaged in the manufacture and distribution of the compound, but more particularly to the public at large (1)." On the basis of research conducted by the Bureau of Mines, U.S. Department of the Interior; the Department of Industrial Medicine, Columbia University; and the Medical College, University of Cincinnati, as well as clinical observation, it was concluded that TEL was a highly toxic industrial compound but that little or nothing was known of the public health effects of its use as a gasoline additive. The Conference thereupon adopted the following resolution:

"It is the sense of this conference that the Surgeon General of the United States Public Health Service appoint a committee of seven recognized authorities in clinical medicine, physiology, and industrial hygiene, to present to him, if possible, by January next, a statement as to the health hazards involved in the retail distribution and general use of TEL gasoline motor fluid; and that this conference indorse as wise the decision of the Ethyl Corp. to discontinue temporarily the sale of ethyl gas; that this investigation shall be paid for exclusively out of public funds; and the results of this investigation shall be reported back to a public conference called for the purpose by the United States Public Health Service, at which labor shall be represented (2)."

To carry out this mandate, the Surgeon General appointed a working committee consisting of the following persons: Dr. A. J. Chesley,

health officer, Minnesota State Board of Health; Dr. William H. Howell, professor of physiology, The Johns Hopkins University; Dr. Reid Hunt, professor of pharmacology, Harvard University; Dr. Walter S. Leathers, professor of preventive medicine, Vanderbilt University; Dr. Julius Stieglitz, professor of chemistry, University of Chicago; and Dr. C. E. A. Winslow, professor of public health, Yale University. This committee first met on June 15, with Dr. Howell serving as chairman. Following delineation of scope by the committee, the Office of Industrial Hygiene and Sanitation was placed in charge of the investigation. The committee completed its mission on January 17, 1926, when it met to draft and sign the final report on the investigation, subsequently issued as Bulletin 163, "The Use of Tetraethyl Lead and Its Relation to Public Health." This bulletin gives the results of clinical and laboratory studies of persons exposed to TEL, findings of atmospheric and dust studies, and suggested recommendations and regulations. The clinical study did not give any decisive indication of lead poisoning among the chauffeurs or workers in garages where ethyl gasoline was used as a motor fuel.

The proposed regulations covered (1) the manufacture of TEL and the blending of the latter to make ethyl fluid, (2) mixing, (3) distribution of ethyl gasoline, and (4) precautions for automobile garages, repair shops, service stations, and filling stations.

The complete proposed regulations are contained in appendix C of this report. They were presented to and approved by the Conference of State and Territorial Health Officers on May 25, 1926. It was proposed that these regulations be adopted by the several States to secure uniformity of control. The restriction on the sale and distribution of ethyl gasoline was removed shortly thereafter.

Following the 1926 Conference of State and Territorial Health Officers, the president of the Ethyl Corp. voluntarily agreed to manufacture, blend, and market TEL and leaded gasoline in accordance with the intent of the recommended regulations. This statement of intent served to establish standarized nation-wide safety procedures and apparently removed the pressure for State regulations. Consequently, no State or Federal laws have been enacted pertaining to the hygienic aspects of TEL. In addition, the recommended regulations were adopted as understandings by many national governments and have provided a basis for world-wide health and safety practices by TEL manufacturers and petroleum refiners.

When the E. I. duPont de Nemours Co. began marketing TEL in 1946, the company advised the Surgeon General that it would abide by the intent of the recommended regulations.

Since 1926 it has been necessary to amend the recommended regulations. These minor revisions have been published in *Public Health*

Reports (vol. 43, Nov. 30, 1928, p. 3147 and vol. 61, Oct. 11, 1946, p. 1487). More recently, a color system for aviation gasoline of various octane ranges was recommended for the U.S. Air Force.

Information furnished by the Bureau of Mines on the analysis of automobile gasoline confirmed the fact that 3.0 cc. gal. is the maximum concentration of TEL being used in commercial gasolines (3).

In July and August 1958, a representative of the Public Health Service visited certain plants of the Ethyl Corp., the Esso Standard Oil Co. of Louisiana, and the DuPont Co. These visits and interviews with company technical and medical officials revealed that very high hygienic standards were being used in the manufacture, transportation, and mixing of TEL and leaded gasoline.

Purpose of Committee

The 1925 committee concluded "that there are at present no good grounds for prohibiting the use of ethyl gasoline of the composition specified as a motor fuel provided that its distribution and uses are controlled by proper regulations." This conclusion was qualified by the statement that, although the study was most careful and conscientous, it was subject to criticism because of the relatively small number of individuals exposed to the effects of ethyl gasoline for a comparatively brief period of time. The committee also noted, "It remains possible that, if the use of leaded gasoline becomes widespread, conditions may arise very different from those studied by us which would render its use more of a hazard than would appear to be the case from this investigation (2)." For these reasons, the committee recommended that the study be continued to determine any potential menace to the health of the general public after prolonged use or under conditions not foreseen at that time.

The recommendation of the committee that the amount of TEL be limited to 1 part in 1,260 parts of gasoline (approximately 3 cc. per gallon of gasoline) was based primarily on commercial practice rather than on hygienic considerations. This concentration has been adhered to by the industry. However, in 1934, after consultation with the Surgeon General, the TEL content of gasoline used solely for aviation purposes was increased to 4.5 cc. per gallon.

On November 26, 1958, Mr. B. B. Turner, president of the Ethyl Corp., in a conference with the Surgeon General, sought the advice and guidance of the Public Health Service on increasing the maximum concentration of tetraethyl lead in automotive gasoline from 3.0 to 4.0 cc. per gallon. Although the Public Health Service has no legal authority to control the amount of TEL in gasoline, the Surgeon General indicated that he would appoint an ad hoc committee to advise whether the proposed increase might represent a public health hazard.

At the request of the Surgeon General the following specialists in occupational health and toxicology were appointed by the Chief of the Occupational Health Program as members of the Ad Hoc Committee on Tetraethyl Lead: Leonard Greenburg, M.D., commissioner, Department of Air Pollution Control, City of New York; H. H. Schrenk, Ph.D., managing director, Industrial Hygiene Foundation, Mellon Institute; J. H. Sterner, M.D., medical director, Eastman Kodak Co.;

W. P. Yant, D.Sc., director of research, Mine Safety Appliances Co.; and Stanley H. Osborn, M.D., commissioner of health, Connecticut State Department of Health, representing the State and Territorial Health Officers' Association.

The Committee met on January 8 and 9, 1959, in Washington, D.C. The meeting was chaired by Dr. H. J. Magnuson, Chief of the Occupational Health Program, Public Health Service, with Mr. H. N. Doyle, Assistant Chief, acting as reporter. The following individuals attended various sessions of the meeting in a liaison or technical capacity: *Ethyl Corp.:* Dr. George Kirby, vice president for research; Dr. Robert A. Kehoe, medical consultant; and Mr. Richard Scales, director of research, Detroit Laboratory. *E. I. duPont de Nemours & Co.:* Mr. J. R. Sabina, assistant planning manager, Division of Petroleum Chemicals, and Dr. John A. Zapp, director, Haskell Laboratory. *Public Health Service:* D. J. Birmingham, M.D., chief, clinical services, Occupational Health Program; H. E. Stokinger, Ph.D., chief, toxicologic services, Occupational Health Program; W. M. Gafafer, D.Sc., technical adviser, Occupational Health Program; V. G. MacKenzie, assistant chief, Division of Sanitary Engineering Services; H. E. Heimann, M.D., assistant chief, Air Pollution Medical Program. *Department of Health, Education, and Welfare:* Sidney Edelman, Assistant Chief, Public Health Division, Office of the General Counsel.

The procedures of the Committee were in accordance with a Department of Justice directive dated October 19, 1950, of which the participants were advised by the chairman. The agenda of the Committee appears as appendix A.

The history of its relationship with the manufacturers of TEL was presented by the Public Health Service; and data on the technical reasons for the increase, the results of certain research studies, the consumption of TEL by year, and a review of the medical problem were presented by representatives of the Ethyl Corp. Representatives of the duPont company made no formal presentation but supplied production figures and information on actual and estimated TEL demand on United States producers and aided in clarifying technical points.

The Committee's report has been transmitted to the E. I. duPont de Nemours Co., the Ethyl Corp., and the American Petroleum Institute, the letter of transmittal appearing as appendix B.

Consumption of Tetraethyl Lead

The domestic consumption of TEL has shown a dramatic increase from 700,000 pounds in 1924, when it was first introduced, to a high of 5.1 million pounds in 1956, with a slight decrease in 1957 and 1958. The estimated domestic TEL consumption is shown in table 1. Table 2 shows the average TEL content of domestic gasoline by year from 1924–58. Table 3 gives the estimated projected consumption of TEL through 1963 based on both a 3- and 4-cc. limit, and table 4 projects the estimated TEL content of gasoline for the period 1959–63 on the same basis. The projections offered by the duPont Co. are slightly lower than those submitted by the Ethyl Corp., both in total demand for TEL and its concentration of gasoline (table 5 and fig. 1). According to data supplied by the duPont Co., lead used in the production of TEL represents 15 percent of the overall United States lead consumption.

TABLE 1. *Estimated domestic tetraethyl lead consumption in millions of pounds, 1924–58*

Year	Automotive	Aviation	Total	Year	Automotive	Aviation	Total
1924	0.7		0.7	1942	77.3	25.2	102.5
1925	.3		.3	1943	90.5	62.5	153.0
1926	1.0		1.0	1944	92.7	129.6	222.3
1927	1.9		1.9	1945	126.6	91.6	218.2
1928	3.1		3.1	1946	134.8	6.4	141.2
1929	8.5		8.5	1947	154	13.9	168.6
1930	14.5		14.5	1948	194	21	215
1931	15.8	0.2	16.0	1949	210	24	234
1932	10.2	.3	10.5	1950	285	27	312
1933	19.1	.4	19.5	1951	291	13	304
1934	34.0	.6	34.6	1952	305	52	357
1935	38.2	.8	39.0	1953	365	78	443
1936	45.2	.7	45.9	1954	388	51	439
1937	56.6	1.2	57.8	1955	432	55	487
1938	59.4	1.5	60.9	1956	451	59	510
1939	79.8	1.5	81.3	1957	440	58	498
1940	113.6	3.1	116.7	1958	396	59	455
1941	138.0	7.4	145.4				

Data supplied by Ethyl Corp. Figures prior to 1945 include both domestic consumption and export. Aviation figures include all United States military use regardless of site of operation.

TABLE 2. *Average tetraethyl lead content of domestic automotive gasoline based on 3-cc. limits, 1924–58*

Year	ml./gal.	Year	ml./gal.
1924	0.03	1942	.92
1925	.01	1943	1.25
1926	.03	1944	1.25
1927	.04	1945	1.48
1928	.06	1946	1.25
1929	.15	1947	1.34
1930	.25	1948	1.53
1931	.26	1949	1.59
1932	.18	1950	2.00
1933	.33	1951	1.69
1934	.55	1952	1.89
1935	.58	1953	2.15
1936	.62	1954	2.22
1937	.72	1955	2.28
1938	.76	1956	2.32
1939	.96	1957	2.22
1940	1.30	1958	1.98
1941	1.40		

Data supplied by the Ethyl Corp.

TABLE 3. *Projected average tetraethyl lead content of domestic automobile gasoline based on 3- and 4-cc. limits, 1959–63*

Year	3 ml.	4 ml.	Year	3 ml.	4 ml.
1959	2.09	2.19	1962	2.60	2.84
1960	2.29	2.40	1963	2.70	3.07
1961	2.46	2.62			

Data supplied by Ethyl Corp.

TABLE 4. *Estimated tetraethyl lead consumption in millions of pounds based on 3- and 4-cc. limits, 1959–63*

Year	Automotive		Aviation	Total, automotive and aviation	
	3 ml.	4 ml.		3 ml.	4 ml.
1959	423	443	59	482	502
1960	477	500	59	536	559
1961	530	565	58	588	623
1962	576	630	56	632	686
1963	615	700	54	669	754

Data supplied by Ethyl Corp.

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TABLE 5. *Tetraethyl lead demand on United States producers, 1958–67*

Year	Domestic motor gasoline			
	3-cc. limit		4-cc. limit	
	TEL conc. cc./gal. pool	TEL demand, millions of lbs.	TEL conc. cc./gal. pool	TEL demand, millions of lbs.
1958	2.00	376	2.00	388
1959	2.10	418	2.10	418
1960	2.10	436	2.13	441
1961	2.20	473	2.23	480
1962	2.20	492	2.26	505
1963	2.25	524	2.30	532
1964	2.25	543	2.39	577
1965	2.25	560	2.42	605
1966	2.25	580	2.52	650
1967	2.25	601	2.55	680

NOTE: 30 percent premium, 70 percent regular ratio assumed.
1958 premium TEL content—2.5 cc. gal.
1958 regular TEL content—1.8 cc. gal.
3-cc. limit 1967 premium TEL content—2.75 cc. gal.
3-cc. limit 1967 regular TEL content—2.05 cc./gal.
4-cc. limit 1967 premium TEL content—3.6 cc. gal.
4-cc. limit 1967 regular TEL content—2.2 cc. gal.
Data supplied by E. I. duPont de Nemours & Co.

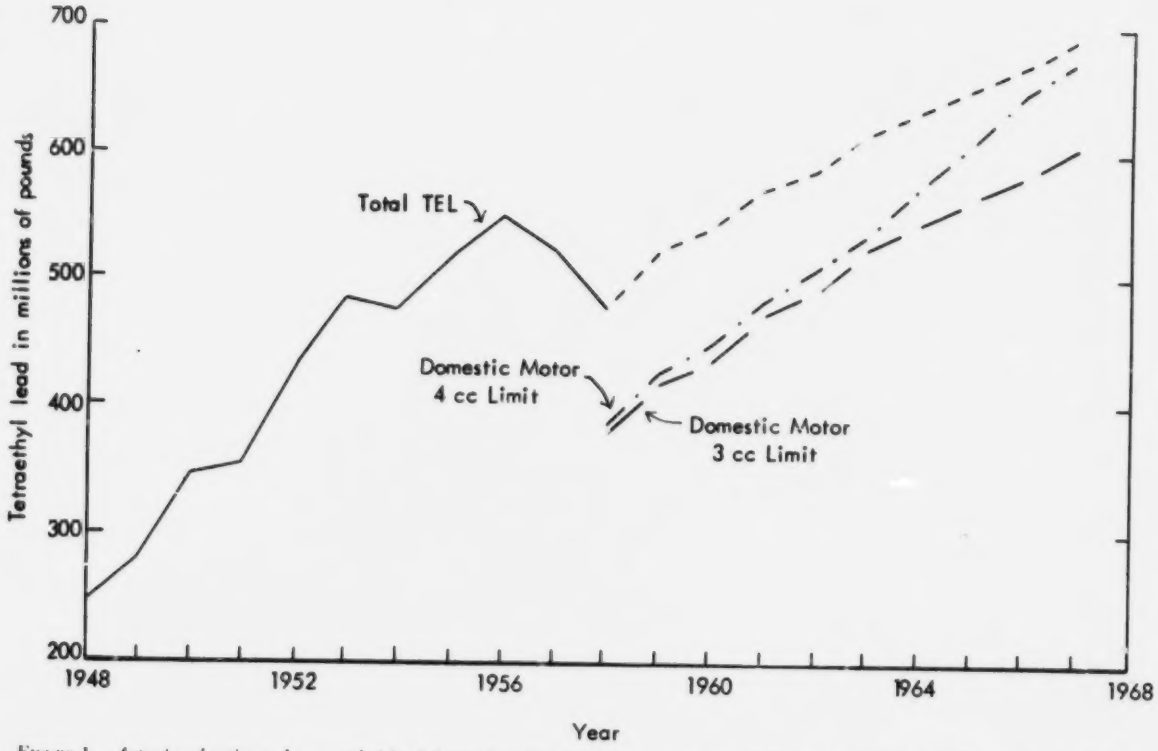

FIGURE 1. *Actual and estimated tretraethyl lead demand on United States producers, 1948–67 (Presented by E. I. duPont de Nemours & Co., Inc.).*

Health and Environmental Data

Public Health Service

The Public Health Service has no knowledge of any occurrence of clinical lead poisoning in the manufacture, blending, mixing, or use of TEL or in the use of leaded gasoline since the adoption of the recommended regulations of the 1926 Committee. It is known, however, that some deaths and clinical cases have resulted from the cleaning of tanks containing leaded gasoline. Although the recommended regulations did not include tank cleaning, this operation has been covered by subsequent safety measures advocated by the manufacturers of TEL and the petroleum companies.

Since the 1925 investigation, there have been no follow-up studies of large population groups on how adding TEL to gasoline affects the total body burden of lead. This information is vitally needed by the committee. It is known, however, that the population is exposed not only to lead discharged by motor vehicles but also to other sources of lead in the atmosphere and that the body accumulates lead through food, water, and tobacco. Research has shown that blood lead concentrations of individuals with no industrial lead exposure range from 0.01 to 0.06 mg. per 100 grams of whole blood and urine lead ranges from 0.01 to 0.08 mg. per liter. Because of the vastly improved microanalytic methods for determining lead, it would be fallacious to compare these values with similar ones obtained before the widespread use of TEL.

The significance of natural sources is suggested by Kehoe's findings of measurable amounts of lead in clinical specimens of a remote Central American tribe (4). A considerable body of information is available on the concentration of lead in the atmospheres of metropolitan areas. A 5-year atmospheric pollution study of Cincinnati, Ohio, revealed atmospheric lead values ranging from less than 0.1 to 11.5 micrograms per cubic meter of air, with an average of 1.2 micrograms in rural areas, 2.0 in residential areas, 3.2 in industrial areas, and 2.7 micrograms for all areas (5). The most comprehensive data come from the National Air Sampling Network of the Public Health Service (6). Information obtained in 15 cities in the United States, shows a range in atmospheric lead values from a high of 9.5 micrograms in Philadelphia to a low of 0.5 microgram per cubic meter in Minneapolis (table 6). Since the sampling site varied from city to

city and was seldom in areas of heaviest traffic or industry, these values are not comparable, nor do they necessarily represent maximum concentrations.

TABLE 6. *Particulate atmospheric lead concentrations in selected cities*

City and population size	Average values, micrograms per cubic meter	City and population size	Average values, micrograms per cubic meter
Over 2,000,000:		From 500,000 to 2,000,000—Continued	
Los Angeles	5.2	Portland	1.2
Detroit	2.9	Atlanta	1.8
Philadelphia	9.5	San Francisco	2.4
Chicago	2.8	Minneapolis	.5
New York	2.8	Less than 500,000:	
From 500,000 to 2,000,000:		Fort Worth	.8
Cincinnati	1.6	Charleston	1.1
Kansas City	1.0	Louisville	.6

Based on tables in reference 7.

Values obtained in nonurban areas in the vicinity of certain cities range from 0.1 to 0.9 microgram per cubic meter. A more recent report of the Public Health Service gives maximum, minimum, average lead concentrations and monthly averages in 50 cities participating in the national Air Sampling Network. Average values ranged from 1 to 57 micrograms of lead per cubic meter and pronounced seasonal variations are apparent (6).

The generally accepted maximum allowable concentration for industrial exposure based on an 8-hour workday is 200 micrograms per cubic meter.

Technical Aspects

Statement Presented by Dr. George F. Kirby, Vice President, Research and Development, Ethyl Corp.

I—Scope and Conclusions

Since 1926, the concentration of tetraethyl lead (TEL) in motor gasoline has been limited to 3 ml. per gallon. This limit was established following a report of an advisory committee convened by the Surgeon General of the Public Health Service.

As a result of technical and economic developments during the intervening 32 years, many refiners are now approaching the 3 ml./gal. limit and would benefit from an upward revision in the permissible concentration.

The present committee will consider whether such an upward revision of the TEL limit is in order. Room for such an inquiry is left by the basis on which the 3 ml. TEL limit was originally chosen. Specifically, this level was selected primarily for technical rather than hygienic reasons and is based on considerations which are largely superseded by more recent technical knowledge and experience.

Through intensive work in the area of the committee's field of inquiry, a considerable body of technical and economic data has been accumulated pertaining to the need for raising the TEL limit. These data are presented in section II.

On the basis of these data, we have reached the conclusion that an increase in TEL concentration above 3 ml. will have the following *economic effects:*

(1) A saving of our national crude oil resources
(2) Decrease in future capital requirements by oil refiners
(3) Improved ability of many refiners to meet competition
(4) Economic advantages to the motoring public

During the past few years, we have carried out technical investigations on the *safety aspects* of the proposed increase, and these findings are reported in section III. From them we have drawn the following conclusions:

(1) The use of 4 ml. per gallon is technically sound from the point of view of engine durability.
(2) The composition and particle size of particulate lead compounds exhausted from automobiles is not changed by going from 3 ml. to 4 ml. per gallon.

(3) The presence of TEL in motor gasolines does not influence smog formation.

(4) The presence of TEL in motor gasolines does not influence the level of eye irritation, aldehydes concentration, and rubber cracking due to contact with exhaust gases.

Accordingly, it is our conclusion that approval of a 4 ml. gallon TEL concentration in automotive fuels will have a favorable effect on the national economy without creating a public health hazard or a technical problem in the areas discussed herein.

It will be the purpose of this statement to provide information on which the foregoing conclusions are based. No attempt is made herein to reach conclusions on medical aspects involved in an increase of TEL limit from 3 ml. to 4 ml. gal. This phase is covered in a separate report submitted by Dr. Robert A. Kehoe.

II—Economic Aspects of TEL Concentration

Technologic Need for TEL

Antiknock requirements in gasoline.—The efficiency of a gasoline engine depends primarily upon its compression ratio. By definition, this is the ratio of the volumes inside the combustion chamber at the end and beginning of one piston stroke. The higher the compression ratio, the more efficient is the engine in terms of power and fuel economy.

Figure 2 shows typical improvements which can be achieved in fuel economy as the compression ratio of an engine is increased. Thus, at current and foreseeable compression ratios, an increase in compression ratio from 8:1 to 14.5:1 will increase mileage per gallon by 30 percent if other conditions remain equal. No matter what the size or weight of the automobile and no matter what its equipment, its gasoline consumption will be less by virtue of a higher compression ratio.

As a result of these advantages, there has been a steady trend toward increased engine compression ratios. This is illustrated by figure 3 which shows that the average compression ratio of passenger cars in production rose from 4.4 in 1925 to approximately 9.5 at the present time.

As the compression ratio in the engine is increased, there is a tendency of the fuel to cause a knocking. Such knocking is objectionable because it is a waste of power and can be harmful to the engine. However, it can be eliminated by using gasoline of improved quality.

Thus, the highly efficient automotive engines in public use today have been made possible by two simultaneous major efforts: The development of high-compression engines and the development of high-quality gasolines which can be used satisfactorily in these engines.

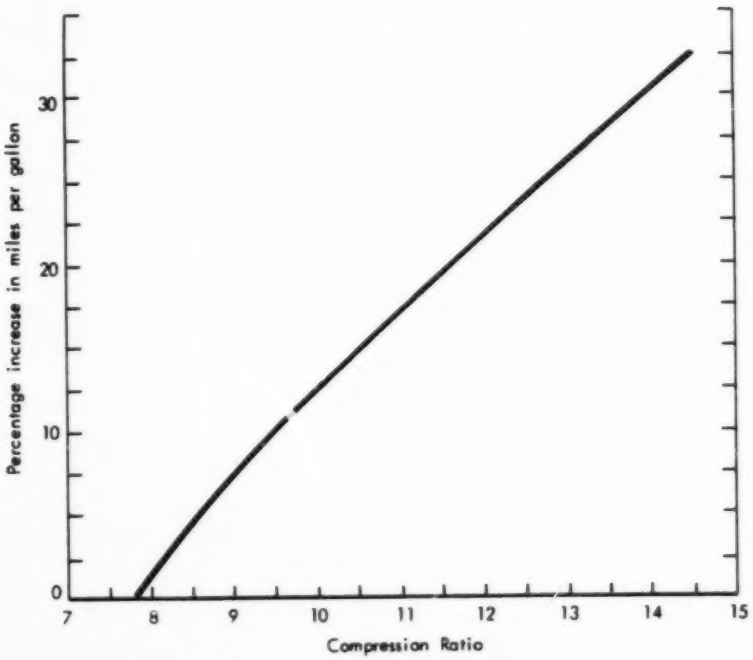

FIGURE 2. *Miles per gallon in relation to compression ratio.*

In this connection, the key criterion of fuel quality is the antiknock value which is measured in terms of octane number.[1] Figure 4 illustrates how the average octane numbers of regular and premium grade gasolines have risen steadily since 1925, a trend which still continues.

Methods for increasing octane numbers.—The refiner has at his disposal two methods for increasing the octane number of his gasoline. One of these is the addition of TEL, which is the most effective known antiknock agent. The other approach calls for upgrading by means of refinery processes.

Until about 8 years ago the processes developed by the refining industry to increase fuel antiknock quality were such that they also increased the amount of gasoline obtained from each barrel of crude oil processed. The main processes in this category are thermal and catalytic cracking of gas oil and middle distillates obtained from petroleum. These techniques, however, have about reached their limit in improving antiknock quality.

Therefore, refiners now find it necessary to utilize reforming processes. In these newer processes, gasoline of intermediate quality is

[1] "Octane number" of a test gasoline is defined as the percent isooctane in a mixture of isooctane and n heptane which has the same antiknock characteristics as the test gasoline. A high octane number denotes high antiknock quality of gasoline.

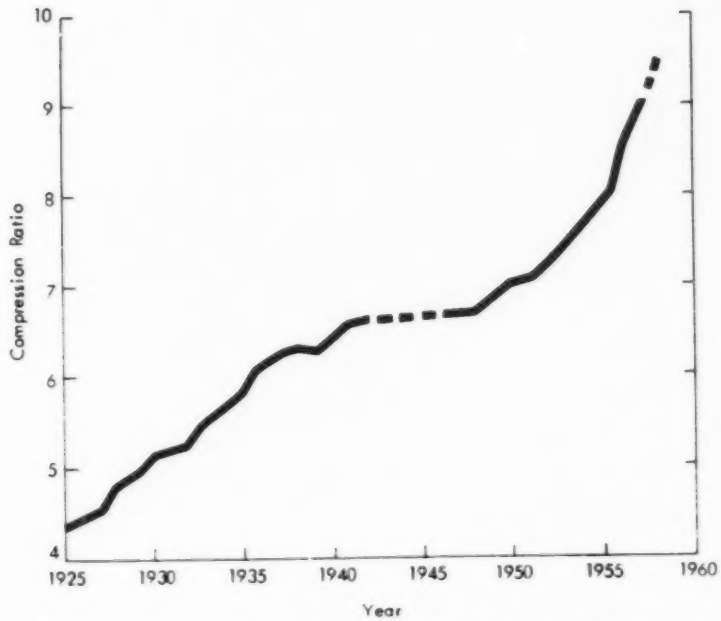

FIGURE 3. *Average compression ratio of passenger cars in production since 1925.*

the feedstock. In the course of processing, the antiknock value is raised but some of the gasoline is converted to other, less valuable products. As a result, the amount of gasoline recovered per barrel of crude oil is decreased.

It is estimated that the industry today reprocesses about 1.5 million barrels of gasoline per day in this manner to meet current antiknock quality requirements. This treatment is accompanied by a loss in yield amounting from 10 to 20 percent of the gasoline reprocessed. The problem is compounded because the industry is already using almost the entire available supply of feedstocks which are most suitable for this type of processing.

In almost every instance, octane improvement is accomplished by a combination of TEL addition and processing methods. The balance between these two methods for achieving octane improvement is struck by the refiner with a view to the most favorable overall economies.

In this picture, the use of TEL offers the refiner the following economic advantages:

(1) Lower consumption of crude oil per gallon of gasoline produced, since further upgrading of gasoline by reforming methods causes a decrease in yield. Present level of TEL

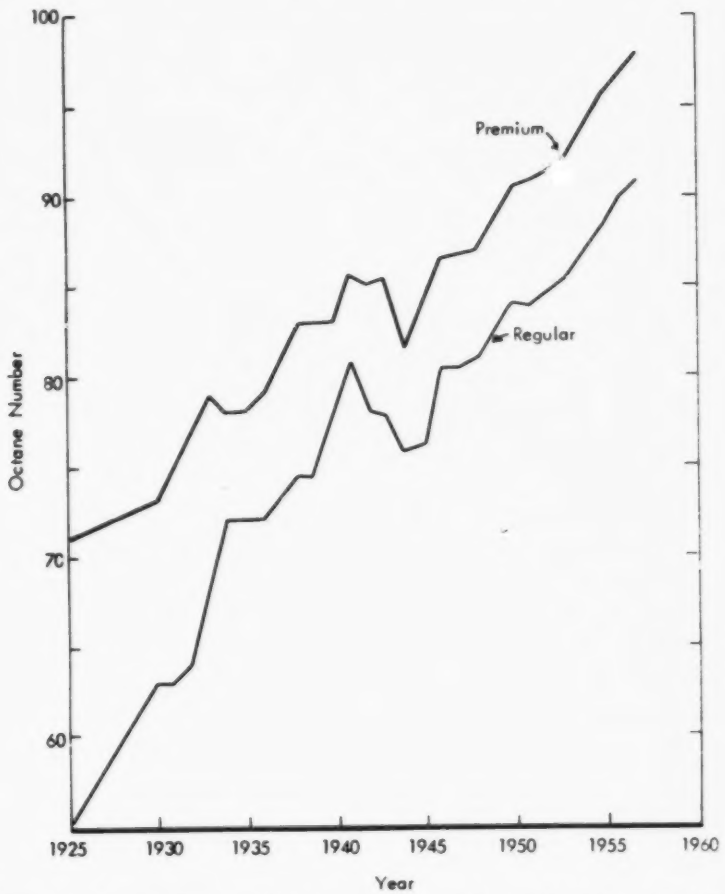

FIGURE 4. *Average octane numbers of regular and premium grade gasolines since 1925.*

usage saves an estimated 250 million barrels of crude oil each year in this manner.

(2) Reduced capital requirements since equivalent processing facilities need not be built. In this manner present level of TEL usage has saved an investment estimated at more than $1 billion.

These benefits associated with current TEL use can be extended as the approved concentration limit is increased.

Restrictiveness of Present TEL Limit

The figures cited in the preceding section concern the United States economy as a whole. It should be borne in mind, however, that the

amount of TEL used in gasoline varies from refiner to refiner and from day to day, just as variations occur in the unleaded gasolines.

While the overall average concentration of TEL in motor gasoline is currently 2.0 ml. per gallon, there are a great many individual refiners whose particular requirements have made it necessary for them to approach the present limit of 3 ml. per gallon, especially in the premium grade fuel. In 1958, gasoline containing between 2.5 and 3.0 ml. TEL accounted for 33 percent of all gasoline and 67 percent of all premium gasoline produced.

Refiners operating in this range of TEL content are deprived of day-by-day operating flexibility, and they face a difficult economic competitive situation as the octane number of marketable gasoline continues to increase.

That such a continuing rise in octane number will develop is our definite expectation. Our present estimates indicate that increase in antiknock quality will continue until the octane number reaches 100 for regular grade fuels and about 105 for premiums. This compares with octane numbers of 92 and 99 at the present time.

With such a trend in the offing, it is certain that the number of refiners approaching the 3 ml. TEL limit will increase rapidly. Our current forecast anticipates a national average TEL concentration of 2.7 ml. by 1963.

Advantages of Increased TEL Concentration

With so many refiners approaching the 3 ml. limit and with continuing upward pressure on the octane number of gasoline, we have found considerable interest on the part of refiners in the possibility of using TEL in excess of the present limit. From the national point of view, we can see four significant benefits which an increase in the TEL limit can achieve:

(1) The more extensive use of TEL will preserve our national crude oil resources since it will minimize the amount lost in severe upgrading processes.

(2) Where the use of higher concentration of TEL is economically the most attractive route, it will have the effect of decreasing capital requirements by oil refiners for new process installations.

(3) Most particularly, accessibility of higher TEL concentrations would be of specific advantage to many refiners who are unable or find it untimely to raise capital for process equipment that they would otherwise need to remain competitive in antiknock quality.

(4) The overall effect will be the availability of a more economic source of gasoline to the motoring public.

III—Safety Aspects of TEL Limits

Basis of Present 3 ml. per Gallon Limit

Before revision of the presently established limit on TEL concentration can be considered, it is necessary to review the basis on which this limit was established in 1926, and to determine whether this basis is still valid in 1959.

When TEL was first introduced in 1923, 3 ml. per gallon TEL was felt to be the highest concentration justifiable on a cost effectiveness basis. This choice was subsequently affirmed by the Surgeon General's committee as a safe hygienic condition and was established as a limit. There is, however, nothing in the underlying work which would suggest that 4 ml. per gallon is a harmful concentration; indeed, there was no valid reason at that time for testing such higher concentrations.

The technology situation which led to the establishment of the 3 ml. limit in the 1920's took account of the types of gasoline then in common use, of their response to TEL, and of the relatively high price of TEL which prevailed at the time (1.0 cent per ml. vs. about 0.2 cent today). Further account had to be taken at the time of the adverse effect of high-TEL gasolines on the durability of the automobile engines of that era.

The technologic basis which controlled selection of the 3 ml. per gallon TEL limit in the 1920's is no longer valid today. Refining methods today differ from processes then in use. The resulting gasolines are of different character and lead response, TEL price has been greatly reduced, and modern engines are well able to cope with gasolines containing 4 ml. TEL.

Advisability of Raising the TEL Limit of 4 ml. per Gallon

Two key considerations made us investigate the advisability of raising the TEL limit above 3 ml. per gallon.

(1) The economic need which is developing for such an increase as a result of modern technology, and

(2) The realization that the basis on which the present limit was selected does not preclude consideration of higher concentration, since 3 ml. per gallon had not been established as an upper limit from the point of view of public safety.

We have felt strongly that a change in allowable TEL limit can be considered seriously only if no adverse effect whatever would result to the public interest and health. We have therefore carried out extensive tests and studies designed to answer the following questions:

(1) Is the use of fuel containing 4 ml. TEL per gallon technically sound in today's automobile engines?

(2) Can the upper limit of TEL concentration be raised above 3 ml. per gallon safely, that is, without resulting in a hazard

to public health and welfare through any means, including lead poisoning, smog formation, or formation of irritants?

Our studies indicate an affirmative answer to both of these questions. Our technical findings bearing on these points are summarized below.

Effect of 4th milliliter TEL on engine durability.—As suppliers of fuel additives to the petroleum industry, we felt that we had to assure ourselves that increasing the amount of TEL in gasoline would not be harmful to engines.

To determine this, our laboratories have carried out an extensive investigation of the engine effect of gasolines containing 4 ml. TEL compared to the same gasolines containing 3 ml. These tests were carried out in passenger cars, trucks, farm tractors, stationary engines, and other equipment. The results of these studies have convinced us that, in the utilization of fuels containing 4 ml. per gallon TEL, there will be no undesirable effects with regard to the durability aspects or the deposits built up within the combustion chamber of the modern gasoline engine.

Effect of 4th ml. TEL on particulate lead exhausted from engine.—Ethyl Corp. has carried out intensive investigations on the character of particulate lead exhausted from engines which are run on TEL-containing gasolines.

Tests were carried out in which engines were run under different conditions. All the exhaust gases were passed through an electrostatic precipitator which collected the particulate matter for later study. Techniques, equipment, procedure, and results of this investigation were reported to the American Chemical Society in September 1956. This paper ("Particulate Lead Compounds in Automobile Exhaust Gas" by Hirschler et al.) appears in *Industrial and Engineering Chemistry*, vol. 49, pp. 1131–1142, July 1957.

Briefly, this work indicates that, when vehicles are operated under conditions approximating those of city driving, from 20 to 60 percent of the lead entering the engine is emitted to the atmosphere as particulate matter. The balance of the lead either accumulates in the engine, the lubricating oil, or the exhaust system of the vehicle or is emitted during high-speed driving or under conditions of heavy acceleration. High-speed driving conditions, which are responsible for the greatest lead emission, are, of course, encountered mostly in rural areas.

The studies were so carried out that we could not only investigate the total amount of lead being emitted but also determine the particle size distribution. It was found that, under city driving conditions, about 27 percent of the lead emitted would be present in particulates in excess of 5 microns in diameter. These particles would be expected to settle rapidly and not remain airborne. Under heavier operating

conditions (corresponding to driving on open highways) this percentage was found to increase to about 40 percent of the total lead emitted.

It was also found that TEL concentration is linearly related to the total amount of particulate lead emitted and that it has no influence on the particle size distribution or composition of the exhausted lead. Therefore, we would estimate that the increase in the amount of airborne lead in the atmosphere resulting from automotive use of TEL would be essentially in direct proportion to the increase in average TEL concentration.

Effect of TEL on formation of atmospheric smog and irritants.—With the growing concern over smog, we also felt it necessary to determine whether or not TEL in gasoline could be a factor. Accordingly, we sponsored an extensive study at the Midwest Research Institute, Kansas City. Results are published in *Industrial and Engineering Chemistry*, vol. 50, pp. 673–676, April 1958.

For the study, the laboratories used two large smog chambers, each of about 2,200 cubic feet capacity. In a series of tests, the chambers were operated under air conditions typical of the Los Angeles atmosphere during severe inversion. Under such conditions, simultaneous introduction of automobile exhaust and of sunlight into the chamber caused typical smog symptoms.

To determine whether TEL is a contributory factor to this smog-forming tendency, a statistically-designed series of experiments compared the emission from a lead-free premium gasoline with exhaust from the same gasoline containing 3 ml. of TEL with two different scavenger compositions. TEL used commercially is mixed with "scavengers" which have the purpose of removing lead deposits from the engine. The commercial scavengers are ethylene dibromide and ethylene dichloride. One of the scavengers used in the test corresponded to the standard "Motor Mix" which is by far the most widely used form in which TEL is added to motor gasoline. The other scavenger compositions correspond to the commercially available "Motor Plus" in which the ethylene dibromide concentration is about 20 percent higher.

The tests revealed that the addition of TEL, regardless of scavenger concentration, is without significant influence on the occurrence of such substances as unburned hydrocarbons, nitrogen oxides, oxidant concentration, eye irritants, aldehydes, and rubber cracking agents. These materials are usually considered key factors in smog formation.

Specifically, figure 5 shows that there is no significant difference among the three fuels (leaded and unleaded) insofar as unburned hydrocarbons, nitrogen oxides, and oxidant concentration are concerned. The dotted lines show the 95 percent confidence limits.

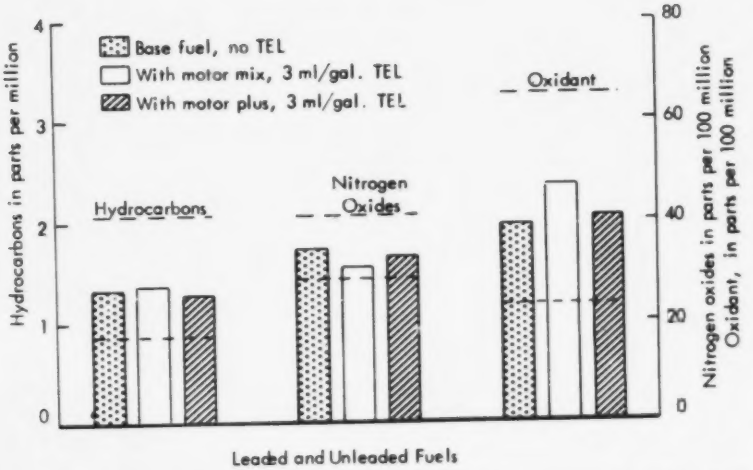
Base fuel, no TEL
With motor mix, 3 ml/gal. TEL
With motor plus, 3 ml/gal. TEL
Hydrocarbons
Nitrogen Oxides
Oxidant
Hydrocarbons in parts per million
Nitrogen oxides in parts per 100 million
Oxidant, in parts per 100 million
Leaded and Unleaded Fuels

FIGURE 5. *Effects of tetraethyl lead and fluid mixes with reference to hydrocarbons, nitrogen oxides, and oxidant (dashed lines indicate 95 percent confidence limits).*

Similarly, figure 6 shows that there is no significant difference among the three fuels as to eye irritation, aldehydes concentration, and rubber cracking effect.

These studies were confined to TEL concentrations of 3 ml. Nevertheless, the observation that 3 ml. TEL has no effect on the occurrence of smog formers and certain nuisance products leads us to expect that no serious problem will arise in these respects by increasing TEL concentration to 4 ml. per gallon.

Effect of Raised Limit on Actual TEL Concentration

In discussing the restrictiveness of the present TEL limit, we have pointed out that the present United States *average* TEL concentration is 2.0 ml., far short of the 3 ml. limit. The immediate need for a higher approved limit is faced by *individual* refiners, whose number may be expected to increase in the near future.

With so many refiners still below the presently approved maximum, it follows that an increase in the approved TEL concentration will not cause an overnight jump in the national average TEL content. Instead, we estimate that future development in national average TEL concentration will follow the trends shown in figure 7. It will be noted that a 1 milliliter increase in approved TEL limit will raise the average concentration in 1962 by only 0.2 ml. By 1963, we expect that the average concentration will have been boosted by 0.4 cc. or 14 percent. It will be during that year, incidentally, that the national average would reach the presently approved limit of 3 ml. per gallon.

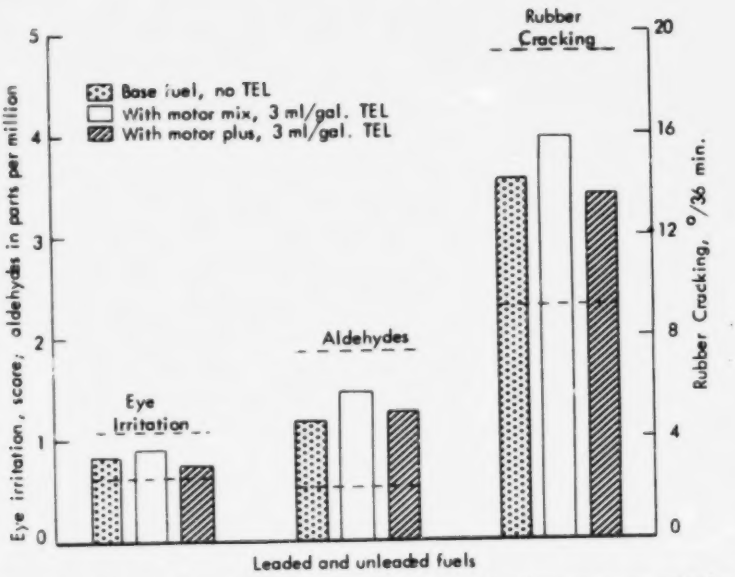

FIGURE 6. *Effects of tetraethyl lead and fluid mixes with reference to eye irritation, aldehydes, and rubber cracking (dashed lines indicate 95 percent confidence limits).*

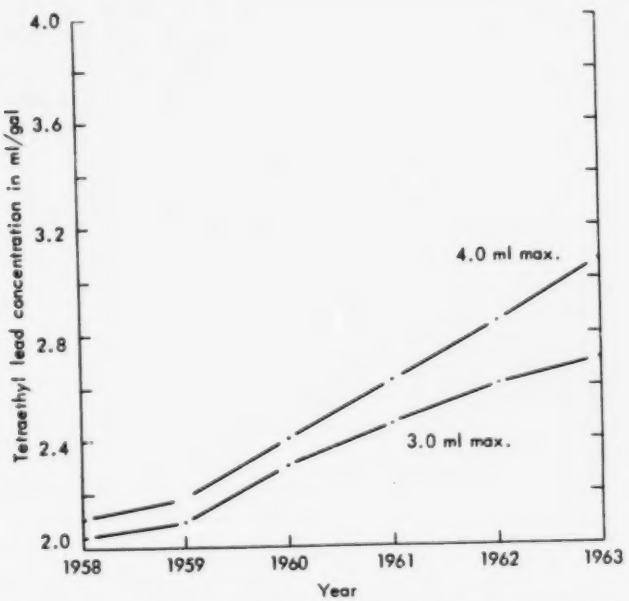

FIGURE 7. *Estimated annual average tetraethyl lead concentrations for United States pool gasoline, 1958–63.*

A national average is not representative of the regional averages, since the amount of TEL used is not the same in different parts of the country. To give this committee a more detailed picture of the probable effect of raising the TEL limit, table 7 provides a geographic breakdown of anticipated increases in actual regional TEL concentration in 1963.

IV—Summary

In summary, we list the various technical factors which appear to be pertinent in considering the possibility of increasing the approved TEL limit from 3 ml. to 4 ml. per gallon.

1. Octane numbers of gasoline will continue to increase, with economic advantages to the motoring public.

2. These additional octane numbers could be supplied in part by the greater use of TEL antiknock compounds than is now possible, at a saving in operating cost, capital expenditures, and natural petroleum resources.

3. The 3 ml. limitation was originally chosen for technical rather than hygienic reasons. The technical situation presents a changed picture today.

4. According to our investigations, the use of 4 ml. TEL per gallon is technically sound from an engine durability standpoint.

5. A broad study of particulate lead compounds exhausted from automobiles shows that the composition and particle size are not influenced by going from 3 ml. to 4 ml. TEL per gallon gasoline.

6. Studies carried out by Midwest Research Institute in smog chambers indicate that exhausted lead does not influence smog.

7. If the limitation in TEL usage is raised from 3 ml. to 4 ml. per gallon, the increase in actual TEL concentration in gasoline will occur gradually.

TABLE 7. *Anticipated increase in tetraethyl lead concentration by geographic region, 1963*

Geographic region	TEL concentration present estimate for 1963	Increased usage for 4th ml.	Percent increase over present estimate for 1963 due to 4th ml.
East	2.82	0.62	22
Midwest	2.75	.44	16
Gulf Coast	2.64	.17	6
West Coast	2.69	.53	20
Rocky Mountain	2.54	.46	18
All United States	2.70	.37	14

Additional Technical Data Supplied by Ethyl Corp.

At the request of various members of the Committee, additional information on specific questions was supplied by the Ethyl Corp. in the following four memoranda.

ETHYL CORPORATION RESEARCH LABORATORIES

Memorandum

JANUARY 14, 1959.

Subject: Lead Concentration in Undiluted Exhaust Gas

The amount of lead exhausted by cars driven on a test cycle typical of city-type service has been found in our studies to range between 20 and 60 percent of the lead burned. Values in the lower range prevail when the car exhaust system is fairly clean, such as in a new car or in a car which has been driven in high-speed highway service prior to city-type driving. Values in the higher range prevail after a car has been driven for several thousand miles in low-speed, city-type service. Based on these results, a reasonable estimate is that the typical city-driven car exhausts half of the lead burned.

To express these values as concentrations of lead in undiluted exhaust gas, the volume of gas produced by a given quantity of fuel and tetraethyl lead must be known. Gas volumes were not measured directly in our exhausted-lead studies because composite samples of lead were collected during a variety of transient and steady-state conditions, and gas volumes were rapidly changing. However, an average gas volume per unit of lead burned can be computed readily and used to establish approximate average concentrations of lead.

At an air-fuel weight ratio of 14 to 1, which is about average for the city-driving test cycle, 1 pound of fuel burns to produce 191 standard cubic feet of exhaust gas. One pound of fuel at a TEL concentration of 3 ml. per gallon also contains 0.511 gram of lead. Therefore, if 50 percent of the lead burned is exhausted with the gas, the gas contains 1.34 milligrams of lead per cubic foot.

This value may be refined further to include lead in specific particle size ranges. Particle size distribution of exhausted lead in two makes of cars was found to be as follows during city-type driving tests:

Weight percent of exhausted lead in particle size ranges

Particle size in microns	Car B	Car M	Average	Particle size in microns	Car B	Car M	Average
	Percent	*Percent*	*Percent*		*Percent*	*Percent*	*Percent*
0-1	3.6	3.7	3.7	5-10	1.1	1.8	1.4
1-2	15.9	15.1	15.5	More than 10	21.4	30.6	26.0
2-3	18.9	16.5	17.7	Unclassified	12.4	11.0	11.7
3-4	16.2	14.4	13.8				
4-5	10.5	6.9	8.7		100.0	100.0	100.0

These values, applied to lead concentrations in exhaust gas, produce the following values:

Lead concentration of exhaust gas containing 50 percent of the lead burned in the fuel

Total lead (all sizes)	1.34 milligrams lead/cu. ft.
5 microns and smaller	0.79 milligrams lead/cu. ft.
5 microns and smaller, plus unclassified lead	0.97 milligrams lead/cu. ft.

(Signed) D. A. HIRSCHLER.

ETHYL CORPORATION RESEARCH LABORATORIES

Memorandum

JANUARY 14, 1959.

Subject: Lead Retained by Automobiles

As a part of our study of exhausted lead, an examination was made of two passenger cars to determine the amount of lead retained by the cars and the location of this lead. The same two cars also had been used in exhausted-lead studies and were exposed both to city-type driving and to high-speed, severe driving conditions. Since high-speed driving tends to expel deposits of lead which accumulate during city-type driving, the lead retained by the cars undoubtedly is less than would be found in cars driven only under mild, city-type conditions. Data are as follows:

Lead retained by automobile exhaust system and lubricating oil

Item	Car make B		Car make M	
Test mileage	26,996		19,345	
Total lead burned, g	5,300		4,154	
	Grams	Percent of input	Grams	Percent of input
Lead recovered:				
Cyl. heads and combustion chambers	76	1.4	117	2.8
Manifolds and exhaust pipes	64	1.2	100	2.4
Mufflers	247	4.7	379	9.1
Tailpipes	5	.1	14	.3
Oil sludge	59	1.1	100	2.4
Oil filters	515	9.7	(1)	
Oil changes	160	3.0	436	10.5
Total retention	1,126	21.2	1,143	27.5

[1] None used.

The results show that car B exhausted 78.8 percent of the lead burned during 26,996 miles, while car M exhausted 72.5 percent of the lead burned during 19,345 miles. These values are consistent with the corresponding studies of exhausted lead. Measurements of exhausted lead showed the cars to exhaust from 20 to 60 percent of the lead burned during city-type driving. Under these conditions the

rest of the lead remains in the car, with part of it forming deposits in the exhaust system. During subsequent full-throttle accelerations or high-speed driving conditions, larger quantities of lead are exhausted, and some of the previously-formed exhaust system deposits are discharged. Since the test cars had been subjected to both types of service prior to analysis of exhaust system deposits, their lead retention appears to be in line with expectations. These studies are presented in more detail in "Particulate Lead Compounds in Automobile Exhaust Gas," by D. A. Hirschler, L. F. Gilbert, F. W. Lamb, and L. M. Niebylski (*Industrial and Engineering Chemistry*, vol. 49, pp. 1131–1142, July 1957).

(Signed) D. A. HIRSCHLER.

ETHYL CORPORATION RESEARCH LABORATORIES

Memorandum

JANUARY 14, 1959.

Subject: The Influence of Compression Ratio and Fuel Tetraethyl Lead Content on Oxides of Nitrogen Concentration in Automotive Exhaust

At the temperatures existing within the combustion chamber of an internal combustion engine, oxides of nitrogen are formed by the fixation of atmospheric nitrogen. The amount of oxygen available within the combustion chamber and the temperature attained are the major factors controlling the degree of fixation.

Since increasing compression ratio results in increasing the peak temperature of the working medium, more oxides of nitrogen will be found at higher compression ratios if there is excess oxygen available. In figure 8 from "Oxides of Nitrogen in Engine Exhaust Gas" by Gilbert, Hirschler, and Getoor, presented before American Chemical Society, September 9, 1957, it can be seen that for an increase in compression ratios of one unit, from 7:1 to 8:1, the concentration of oxides of nitrogen was increased about 8 percent at an air-fuel ratio of 17.0 to 1 where there was ample excess oxygen available. At 12.0 to 1 air-fuel ratio, however, the increased temperatures resulting from increasing compression ratio had no influence on oxides of nitrogen since there was no oxygen available for increased fixation.

The presence of tetraethyl lead in a fuel or its concentration will not influence the oxides of nitrogen concentration so long as the fuel, either clear or containing tetraethyl lead, is of sufficient antiknock quality to prevent knock. Under conditions where the addition of tetraethyl lead to a fuel raises its antiknock quality sufficiently to prevent knock, the tetraethyl lead will reduce oxides of nitrogen by virtue of the reduction in temperature obtained by eliminating knock.

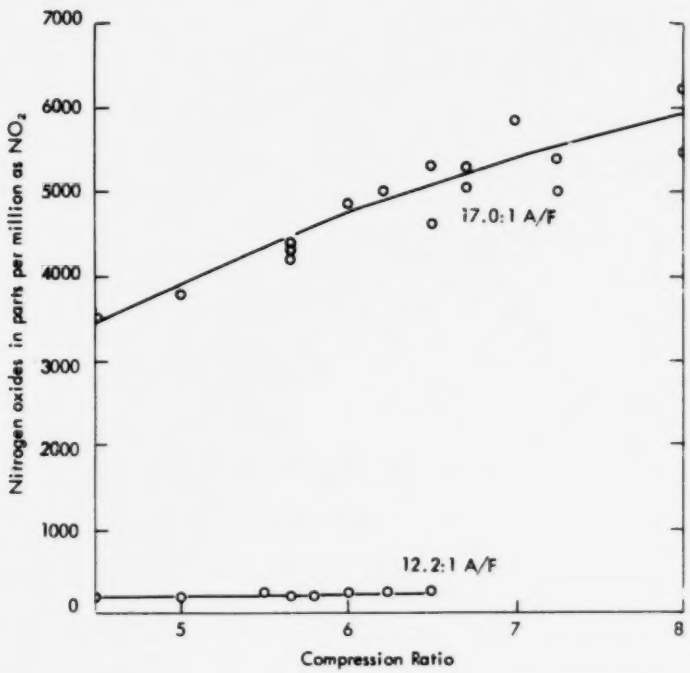

FIGURE 8. *Effects of compression ratio and air-fuel ratio on concentrations of exhausted oxides of nitrogen (CFR engine).*

Figure 9 from the paper referred to above gives data from a multicylinder engine substantiating these statements. The upper, dashed curve indicates oxides of nitrogen concentrations measured at various degrees of knock intensity with a 50-octane number reference fuel. The open circles on the lower curve show a reduction of up to 15 percent in oxides of nitrogen at comparable operating conditions when tetraethyl lead is added to the fuel to eliminate knock. A comparison of the open and closed circles on the lower, solid curve indicates that the concentrations of oxides of nitrogen are identical under nonknocking conditions when an unleaded fuel is compared with one containing tetraethyl lead.

In summary, when oxygen is available, increasing the compression ratio will increase oxides of nitrogen. Increasing tetraethyl lead concentration will have no influence on oxides of nitrogen unless the added antiknock quality reduces knock and then the result will be a decrease in concentration.

(Signed) H. E. HESSELBERG.

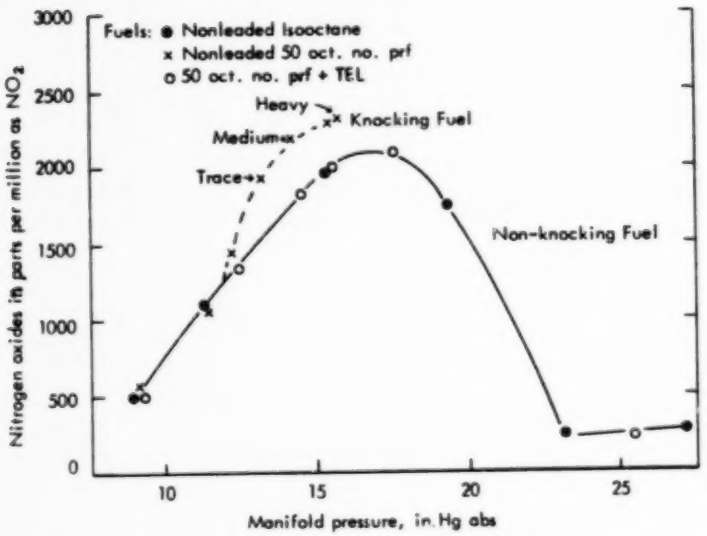

FIGURE 9. *Oxides of nitrogen under knocking and nonknocking conditions (car A: 2,000 engine RPM).*

ETHYL CORPORATION RESEARCH LABORATORIES

Memorandum

JANUARY 14, 1959.

Subject: The Influence of Tetraethyl Lead Concentration on Catalytic Converters

Since the particulate decomposition products of tetraethyl lead must pass with the exhaust gas through any catalytic converter device that would be installed in the exhaust system of a vehicle, a certain percentage of this particulate matter will be deposited on the catalytic material within the converter. In addition to the particulate lead, deposition will accumulate from unburned and partially unburned portions of the fuel and lubricant, and some deposits will occur from the additives in the lubricating oil.

With certain catalytic materials, such as platinum, it is known that the presence of particulate lead very rapidly decreases the catalytic efficiency. No data are available to our knowledge relating the rate of this decrease in efficiency to the tetraethyl lead content of the fuel being used. Since it has been reported that the decrease in activity when tetraethyl lead is present is of a drastic nature, it might be assumed that the relationship between decreased efficiency and tetra-

ethyl lead concentration would be of a logarithmic nature with the first incremental amounts of lead causing the major decrease in efficiency.

With the more recently developed catalytic converter materials, such as the vanadium pentoxide reported by Ford Motor Co., the catalysts appear to be relatively resistant to loss in activity due to the presence of particulate lead. It has been reported that vehicles equipped with such catalytic converters have operated as long as 12,500 miles without suffering serious decrease in catalytic efficiency. This mileage accumulation was on commercially available fuels and it can be assumed safely that the tetraethyl lead concentration ranged from 1.5 to almost 3.0 ml per gallon. Again, no data are available to our knowledge to indicate the effect of fuel tetraethyl lead concentration on the life of catalysts of this type.

The catalytic converter developed by the Oxy-Catalyst Corp. utilizes a catalyst of undisclosed composition, but it appears to be relatively resistant to tetraethyl lead decomposition products. Here again, it has been reported by General Motors Research that satisfactory efficiency has been obtained for 12,500 miles when operated on leaded fuels.

X-ray and electron diffraction analyses in our laboratories on catalysts which have been operated on leaded fuels indicate that reaction occurs between the lead particulate matter and the active catalyst material. This reaction is undoubtedly related to the eventual decrease in catalytic activity. Information is not, however, available which would permit predicting to what degree tetraethyl lead concentration would influence this rate of reaction.

Our laboratories have underway, currently, a cooperative program to evaluate one of the lead-resistant catalytic converters and we will be developing our own data on catalyst efficiency versus mileage accumulation in typical, owner-type operation. We also have underway a program aimed at the development and evaluation of catalytic materials which will have satisfactory conversion efficiency when operated on additive-containing fuels.

(Signed) H. E. Hesselberg.

Hygienic Aspects

Statement by Robert A. Kehoe, M.D., Medical Consultant, Ethyl Corp., March 18, 1959. (Abstracted by H. N. Doyle.) (Full report to be published by Kettering Laboratory, University of Cincinnati, Cincinnati, Ohio.)

Dr. Kehoe's report, an expansion of his remarks to the committee, is divided into two sections: (1) The consideration of occupational health hazards involved under present conditions and under those projected on the basis of an elevated limit; and (2) consideration of the public health hazard involved under present conditions and under those projected on the basis of an elevated concentration of TEL gasoline.

Section 1 treats in considerable detail the hazard to workers involved in the manufacture, blending, transportation, storage, and use of TEL and leaded gasoline. It points out that increasing the concentration of TEL from 3 to 4 milliliters per American gallon would not increase the hazard to the worker under any conditions of use or transportation of TEL or leaded gasoline. The hazard due to manufacture and storage may be magnified due to the larger amounts of TEL involved, but there is no reason to suspect that it could not be controlled.

Since adequate hygienic precautions were instituted in the manufacture, transportation and use of TEL, there have been no significant cases of lead intoxication in exposed personnel. The first warning of a special problem involving the storage of leaded gasoline came in 1928, when a small group of men registered certain unusual complaints of illness following the cleaning of tanks that had been out of service for several months after extended use for storage of leaded gasoline.

By 1940, there had been 80 well-documented cases of poisoning in the continental United States in connection with tank cleaning and repair operations, with about 15 fatalities. It was apparent by this time that drastic measures had to be taken to secure complete compliance with the precautions. Accordingly, the Ethyl Corp. (at that time the only distributor of TEL) decided to use the members of its technical and sales field staff for the supervision of all tank cleaning operations in the United States. Since the institution of these safety precautions [also adopted by the duPont Co.] the incidence of poison-

ing in this country resulting from tank cleaning operations has been essentially nonexistent.

Potential public health problems result from the shipment of cargoes of edible oils and other foodstuffs in tank cars previously used for leaded gasoline, the use of leaded gasoline in gasoline lanterns, and the practice, especially in foreign countries, of using small leaded gasoline containers for household purposes. However, there has been no evidence to indicate that these procedures have endangered the general public. Should these in fact be problems, the increase of 1 cc. per gallon of TEL would hardly intensify the public health problem due to the misuse of leaded gasoline or the transportation of foodstuffs in tank cars which have contained leaded gasoline. This latter practice is discouraged but cannot be entirely prevented, especially in emergency periods when there is a shortage of tank cars.

Section 2 deals with the potential hazard to the general public resulting from the use of lead in internal combustion engines and includes a comprehensive discussion of the metabolism of lead by the human body.

Lead in atmosphere of American cities with special reference to that derived from exhaust of motor vehicles.—TEL is decomposed to inorganic lead compounds in the combustion chambers of internal combustion engines. Under present conditions, a large proportion of the exhausted lead is the double salt, lead chlor-bromide. It is possible that a minute quantity of TEL escapes combustion, but under ordinary circumstances, little opportunity is afforded for TEL to escape from the exhaust system.

The metabolism of lead in an internal combustion engine has been thoroughly studied. It has been determined that a large percentage of the lead accumulates in the motor and exhaust system. Under conditions of high speed driving and rapid acceleration, the internal combustion engine discharges more lead into the atmosphere than it accumulates. The lead discharged under these conditions is in the form of scale and shreds, although some of it is volatilized.

Under usual conditions of city driving, the discharge of lead into the air is low because of the relatively low consumption of fuel, and the condensing effect of the exhaust system. The same is true to a greater or lesser degree when driving in congested areas of a city. Because of the prominence of small size lead particles in the exhaust during slow speed driving, the lead so discharged is readily dissipated by air currents. In whatever dilution it occurs in the breathing zone of man, the lead is respired and absorbed readily.

Source of lead in urban atmospheres.—It appears to have been assumed by many persons and even by some investigators that the lead found in the atmosphere of cities and in the open countryside is derived from the combustion of leaded gasoline. There were few

recorded determinations of atmospheric lead concentrations prior to 1924, when TEL was introduced as a gasoline additive. However, a few early observers, using the then limited methods, found traces of lead and many other elements in the atmosphere. These values were of scientific interest, but of little hygienic importance. Precise measurements of lead in the atmosphere have been made possible only by the technicologic developments of fairly recent years, and only since TEL came into use as a motor fuel. There are many sources of atmospheric lead contamination in addition to TEL. These include the combustion of wood and other vegetation, the finely divided soil in the atmosphere, and dust and fumes from a multiplicity of sources, including the stacks of industry.

The contribution made to atmospheric lead content by the combustion of TEL in automotive fuels has been difficult to determine or even approximate. Measurement of the lead in the air along certain thoroughfares has shown significant quantitative variation correlated with the density of motor traffic, but the actual proportion or quantities derived from motor exhaust cannot be estimated from the data. These statements purport to emphasize the complexity of the problem and to direct discussion to its essential point, namely, that we are and will be concerned with the total quantity of lead in the atmosphere and not solely with the atmospheric lead derived from automobile exhaust.

Quantities of lead in urban atmospheres.—The most comprehensive information on the quantity of lead in urban atmospheres has been obtained in Cincinnati and its environs. The concentration of lead in the atmosphere varies widely from somewhat less than one to more than six micrograms per cubic meter of air. These values increase as one goes from rural zones outside the city to suburban, business and industrial areas. There are also seasonal variations, lower values being obtained in warm weather, and higher values in cold. The average concentration in Cincinnati was somewhat less than 2 micrograms per cubic meter.

Surveys made in other eastern and middle western cities are in harmony with observations made in Cincinnati. Relatively low but not remarkably divergent results have been obtained in Minneapolis and Louisville, while higher but again not grossly divergent values have been observed in such areas as Los Angeles and Philadelphia. Data obtained in Los Angeles and Pasadena in 1954, as well as data gathered from other cities, are especially useful in revealing maximum and minimum values.

Because of the variability of lead in the atmosphere of our cities, as well as other variable circumstances, it is impossible to arrive at any precise estimate of the quantity of lead which is inhaled by the "average" person in the course of an "average" day. It is even more diffi-

cult to estimate the largest quantity to which any person may be exposed in this manner over a period of months without sustaining a significant increase in the lead content of the body. On the basis of certain assumptions, it is possible that the average man may absorb from the atmosphere a quantity of lead in the order of 0.038 milligram per day. Inasmuch as the assumptions made in this calculation are believed to err on the high side, this figure should not be taken too literally. The quantity may be nearer to half this value and other observations to be discussed later indicated that it may not be far from 0.02 milligram per day on the average over a period of years.

General sources of human exposure to lead and approximate contribution made by each source to total exposure.—The average adult North American takes in approximately 0.33 milligram of lead per day in food and beverages. The quantities of lead in the daily food intake vary from somewhat less than 0.1 milligram to 4.0 milligrams, but the mean daily intake is quite stable and the pattern has repeated itself regularly over a considerable period of time. Of this quantity, 0.01 to 0.03 milligram per day are derived from water used for cooking and as a beverage. The remainder is distributed in fairly small quantities over a wide variety of foods and beverages which contain lead from both natural and artificial sources.

The quantities of lead evacuated in the feces of the average North American adult are approximately equivalent to those ingested with his food and beverage. This information, gained in a variety of experimental approaches to the question, indicates that somewhat less than 10 percent of the lead ingested in these small quantities is absorbed. That some alimentary absorption of lead occurs can be demonstrated more readily and more certainly through observations made on the excretion of lead in the urine from day to day. It is evident that the daily urinary output of lead varies with the alimentary intake of lead, and thus it may be ascertained that lead is absorbed regularly from the alimentary tract. Such absorption under normal conditions is nearer to 5 percent of the daily alimentary intake than it is to 10 percent, but there are variations in this respect which would be extremely difficult to describe or explain in detail.

The excretion of lead in the urine of the average normal North American adult amounts to about 0.03 milligram per day when the urinary output averages about 1 liter per day. The lead content of blood of normal healthy American adults is much less variable than that of the urine, ranging from 0.02 milligram to 0.06 milligram per 100 grams of whole blood and averaging somewhat less than 0.03 milligram.

The mean lead content of other body tissues varies widely from 0.01 milligram per 100 grams of fresh muscle tissue to 1.50 milligrams

per 100 grams of rib and long bone. The variation from individual to individual is appreciable but is not remarkable in view of the variations in height and weight, occupations, length of life, and dietary habits. In balanced experiments, normal healthy North American subjects were followed for 13 and 18 months, respectively, during which time the daily intake of lead in food and beverages was determined by the analysis of duplicate samples of everything eaten and drunk and the daily output in the feces and urine. It was found that the output of lead systematically exceeded the intake by about 8 milligrams per year or, by calculation, about 0.02 milligram per day. This metabolic discrepancy whereby the output of lead exceeded the intake, can only be ascribed to a source of lead absorption not accounted for in the design of the balance experiments. This source undoubtedly was the air inhaled by the experimental subject.

The capacity of the tissues for accumulating lead is considerable. If progressive accumulation does not occur under normal conditions, it must be because the human body, once in equilibrium with its environment at a low level of lead intake and absorption, is able to maintain itself in an essentially steady state with reference to its lead content through excretionary loss. The excretionary processes are highly responsive to variations in the absorption of lead, and they appear to be entirely capable of eliminating the regular stream of absorbed lead from day to day so long as this stream varies reasonably within normal limits.

Metabolism of lead under abnormal conditions.—Investigations of occupational conditions and personnel as well as experimental subjects have provided evidence as to the limits of safe levels of exposure on the basis of physiologic criteria. The general rate of urinary excretion of lead by workmen without renal impairment who have been exposed to lead under reasonably uniform occupational conditions for long periods of time gives a valid clue to the severity and hygienic significance of the exposure.

When the mean level of concentration of lead in the urine does not exceed 0.1 milligram per liter, the conditions of exposure are safe or have been within the limits of safe exposure during the period preceding the time of sampling. When this value exceeds 0.15 milligram of lead per liter, occasional cases of lead poisoning may occur in the group. As the excretion level rises above 0.15 milligram per liter, cases of poisoning among the group increase both in number and severity.

The blood may also be used as an index of exposure, but the blood level lags somewhat behind the excretionary level. Experiments have shown that when blood levels do not exceed 0.07 milligram per 100 grams of blood, the occupational conditions are safe or have been within the limits of safety preceding the time of sampling. Blood

lead concentrations in excess of 0.08 milligram per 100 grams indicate that the individual has sustained a dangerous level of lead absorption and retention, and may develop symptoms of lead intoxication. The extent that the level exceeds 0.08 milligram per 100 grams of blood evidences an increasing incidence and severity of lead intoxication.

The rate of urinary excretion of lead increases stepwise over a period of several months until it reaches a peak. It then stabilizes at a level characteristic of the level of lead exposure associated with the occupation. After this stabilization, any significant trend either upward or downward may be regarded as indicative of deteriorating or improving occupational conditions. Lead levels in the range 0.01 to 0.06 milligram per 100 grams of blood are indicative of safe occupational exposures. Levels between 0.06 and 0.08 per 100 grams are abnormal and indicate definite lead absorption but within safe limits. Values above 0.08 milligram per 100 grams point to the necessity of terminating the occupational exposure to lead either temporarily or permanently as the circumstances may dictate.

It is not possible to express the severity of occupational exposures to lead compounds in terms of environmental measurements with anything like the precision represented by the physiologic measurements referred to previously.

The daily addition of a soluble salt of lead of approximately 0.03 milligram (thereby doubling the daily alimentary intake of lead) over a period of a year resulted in a slight but significant and progressive increase in the retention of lead in the body, a very slight progressive increase in the urinary output of lead, but no measurable increase in the concentration of lead in blood. The addition of larger quantities of soluble lead in the diets of three other experimental subjects, to provide a total mean intake of lead of 1.3, 2.3 and 3.3 milligrams per day, resulted in a prompt and progressive increase in the output of urinary lead, which varied directly with the magnitude of the increased lead input. The rates of accumulation of lead in the bodies of the subjects as well as the rate of increase in the concentration of lead in their urine and blood did not diminish as time passed. The increase persisted apparently at an undiminished annual rate for as long as 2 years in one experiment and 4 years in another. However, none of the human subjects sustained any injurious effects or deviation from well-being during any part of the experimental regimen.

It would appear that the absorption of lead from the alimentary tract is within the limits of safety when the average amount available for absorption is on the order of 0.6 milligram per day. It seems likely that an alimentary intake of about twice this amount, while devoid of danger over a period of 4 or 5 years, would not be tolerated without significant probability of an injurious effect over a period of 10 or more years. It appears that about 60 micrograms of lead can

be absorbed daily from the alimentary tract with safety, while the absorption of 120 micrograms per day is potentially dangerous. If this be a reasonably valid conclusion, it follows that an incremental increase in any type of human exposure to lead that would result in the additional absorption of about 30 micrograms of lead per day would be within the limits of safety, while an increase in absorption on the order of 90 or more micrograms per day may be harmful.

To determine the response of human experimental subjects to the inhalation of abnormal quantities of lead, a series of balance experiments have been underway. Healthy human subjects have been followed before, during and after prolonged periods (2 years or more) of exposure in a chamber for 7.5 hours per day, 5 days per week, to air containing dispersed lead sesquioxide in concentrations equivalent to 0.075 milligram (one experiment) and 0.15 milligram (5 experiments) of lead per cubic meter. Mean diameters of the particle size were 0.05, 0.75, and 1.2 microns. When the concentration of lead sesquioxide in the chamber was equivalent to 0.075 milligram of lead per cubic meter of air, the blood stabilized in about 6 months at 0.04 milligram per 100 grams of whole blood. At the exposure level, equivalent of 0.15 milligram of lead per cubic meter, the stabilized level of 0.045 milligram per 100 grams of whole blood was reached after about 8 months. The mean urinary excretion level paralleled that in the blood, levelling off at about 0.06 milligram per liter in the first experiment and at about 0.08 milligram per liter in the second experiment. Thus, it is evident that the 16.5 hours per day of freedom from experimental exposure during 5 days of the week, together with the complete lack of experimental exposure on 2 days, were sufficient to counterbalance the effects of the experimental exposure after some lead had accumulated in the body. An approximate equilibrium between the weekly absorption and weekly excretion of lead was established in the body. The breathing of air containing lead in the concentration of 0.15 milligram per cubic meter with a mean particle diameter of 0.05 micron did not induce a dangerous level of lead absorption in the body of the experimental subject after more than 2 years of exposure. In view of the mean concentration of lead found in the urine and blood of the subject at the termination of the experiment, the margin of safety of the experimental condition was large. The determination of the extent of the alveolar retention of lead under the experimental conditions demonstrated that this was in the order of 35 to 50 percent.

These levels would not simulate the exposure of an urban population to lead derived from the exhaust of automobiles or other sources of atmospheric pollution. The results, therefore, do not define the margin of safety of the general population in terms of any presently known or calculable maximum allowable concentration of lead in the

general urban atmosphere. There can be little question, however, as to the adequancy of the presently accepted standard of safety for respiratory occupational exposure in which the lead compounds are dispersed in a comparable state of subdivision in the atmosphere.

Significance of lead in atmosphere of cities at present time and future outlook with regard to an increase in discharge of lead compounds from exhaust of motor vehicles.—The evidence demonstrates conclusively that the contribution made by the TEL contamination of the atmosphere to the total absorption of lead by the American population has been remarkably small in either relative or absolute terms and that it has been minute physiologically and insignificant hygienically, despite the tremendous increase in the use of TEL. During the past 11 years, during which the greatest expansion of TEL has occurred, there has been no sign that the average individual in the United States has sustained any measurable increase in the concentration of lead in his blood or in the daily output of lead in his urine. In detailed and prolonged study of experimental subjects at the Kettering Laboratory, no evidence has been seen of any quantitative alteration in the normal metabolism of lead, nor has there been any suggestion of an increase in the levels of lead concentration in the urine and blood of normal individuals from many walks of life and many areas of the country.

On the other hand, time is the important factor in the development of a physiologic response to the absorption of minute quantities of lead. It may be that the influence of this factor is yet to be recognized fully in relation to this problem. Should this be true, further investigations will reveal its influence long before any harmful effects will have been induced unless there is a deleterious effect, as yet unsuspected, from prolonged exposure to slightly larger quantities of lead than those present in the natural environment of man. Our concern over the hygienic effect of an increase in the permissive amount of lead in gasoline has been replaced by the assurance that, by applying the methods of investigation that have provided the present background of information, we could establish the effects of the proposed changes in advance of any harmful consequences.

Abstracts of Special Reports Furnished by Ethyl Corporation

1. Oxides of nitrogen in engine exhaust gas by Gilbert, Herschler, and Getoor. Presented at 132nd meeting, American Chemical Society, September 9, 1957.

A study has been made of the influence of engine operating variables on the concentration of oxides of nitrogen present in engine exhaust gas. Total oxides of nitrogen (except N_2O) were measured in exhaust gas samples by a phenoldisulfonic acid method. Single-cylinder engine studies showed that air-fuel ratio, which controls the amount of oxygen in the combustion gases, was the predominant fac-

tor in controlling the concentration of oxides of nitrogen. Lean mixtures (with higher oxygen concentrations) produced high concentrations of nitrogen oxides, which were increased further by such other variables as advanced ignition timing, higher compression ratio and higher intake manifold pressure, which tend to raise peak cycle temperatures. However, with rich mixtures (deficient in oxygen) nitrogen oxide concentrations were low and insensitive to temperature influencing variables. These results imply that the oxides of nitrogen are formed by the fixation of atmospheric nitrogen, with oxygen concentrations and temperatures controlling this process.

In two modern passenger cars of different makes, which were studied over a range of driving conditions, variations in concentrations of oxides of nitrogen generally were related to the manner in which the respective operating conditions affected air-fuel ratio, ignition timing, and intake manifold pressure. At idle, a concentration of only about 37 ppm. was obtained, but under road-load cruise conditions, concentrations increased with higher speed to as high as 3,975 ppm. at 70 mph. During full-throttle accelerations, concentrations were low relative to cruise conditions because of the rich air-fuel mixtures and somewhat retarded ignition normally encountered at full throttle. During moderate accelerations at part-throttle, however, maximum concentrations were exhausted and appear to correspond to the lean mixtures and advanced ignition timing normally found under such conditions. Deceleration produced concentrations of about 100 ppm. and lower. Concentrations were increased by knock by as much as 15 percent, and suppression of knock by either tetraethyl lead or by higher base fuel antiknock quality prevented this increase. Under non-knocking conditions tetraethyl lead had no effect.

2. Particulate lead compounds in automobile exhaust by Hirschler, Gilbert, Lamb, and Niebylski. *Industrial and Engineering Chemistry*, vol. 49, pp. 1131–1142, July 1957.

A comprehensive study has been made of the amount, composition, and particle size distribution of the lead compounds present in the exhaust gases of automobile engines operated on fuels containing tetraethyl lead. The results show that city-type driving tends to exhaust considerably less lead than is burned by the car and that the excess lead which deposits on exhaust system surfaces tends to be exhausted under higher-speed or higher-load operating conditions. The exhausted material ranges in particle diameter from 0.01 micron to several millimeters and is composed mainly of PbCl·Br, alpha and beta forms of $NH_4Cl \cdot 2PbCl \cdot Br$, and $2NH_4Cl \cdot PbCl \cdot Br$. The data provide an explanation for the low lead concentrations which have been reported for urban atmospheres in relation to the consumption of leaded gasolines.

3. Filtration of particulate lead compounds from automobile exhaust gas by Gilbert. Project No. 8U5-M, Research Memorandum No. 209. January 13, 1959.

This exploratory study has shown that it is possible to filter solids from exhaust gases of automobiles with fair efficiency and without a prohibitive pressure drop across the filtering unit. Ceramic fibrous material such as Fiberfrax and Refrasil have shown themselves to be suitable for use as filter beds for periods up to 5,000 miles of general road service. However, the filter efficiencies of this type of filter appear to be too low unless packing density is increased to the point of objectionable back pressures. From the viewpoint of both service life and filter efficiency, a filter unit with the filter media of 18 gage wire nails appears to be much superior to any other unit tested. The primary drawback to a filter of this type, and one which was not studied in this work, is the possible reduction in exhaust gas temperature during the warm-up of such a large mass of metal. If too large a heat loss occurs the unit would interfere with the action of a catalytic muffler, which needs the heat to operate and which should warm up rapidly.

4. Catalytic oxidation of automobile exhaust gases—an evaluation of the Houdry catalyst by Nebel and Bishop. Presented at Society of Automotive Engineers' Annual Meeting, Detroit, Mich., January 12-16, 1959.

Catalytic oxidation of automobile exhaust gases is technically feasible even when using commercial leaded gasoline. Hydrocarbon emission can be substantially reduced by this process. The overall efficiency or percentage reduction depends on many factors, but with the Houdry catalyst a value of 75 percent (after warmup) seems reasonable. Carbon monoxide is oxidized to about the same degree.

It also appears technically feasible to remove substantial amounts of both combustibles and oxides of nitrogen from exhaust gases by combining catalytic oxidation with modified carburetion. With the combination of the Houdry catalyst to remove hydrocarbons and carbon monoxide and the special carburetor to remove oxides of nitrogen, the following reductions (after warmup) seem reasonable: hydrocarbons, 90 percent; carbon monoxide, 75 percent; and oxide of nitrogen, 90 percent.

The Houdry catalyst is resistant to poisoning by tetraethyl lead combustion products. The useful life of the catalyst is estimated to be about 12,000 miles, after which replacement or rejuvenation seems desirable.

There are many difficult problems to be solved before catalytic oxidation can become commercially acceptable. The solution to these problems will require a major engineering development program.

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Appendix A

Agenda Followed at Committee Meeting

DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE, PUBLIC HEALTH SERVICE, BUREAU OF STATE SERVICES, DIVISION OF SPECIAL HEALTH SERVICES, OCCUPATIONAL HEALTH PROGRAM

Ad Hoc Committee to the Surgeon General on Tetraethyl Lead

JANUARY 8 and 9, 1959.

GENERAL SERVICES ADMINISTRATION,
Regional Office Building,
7th and "D" Streets S.W., Room 3652
Washington, D.C.

Subject for discussion: Does the increase from 3 ml. to 4 ml. of tetraethyl lead per gallon of gasoline represent a public health hazard?

Agenda

1. Introduction: Dr. Magnuson.
2. Background: Mr. Doyle.
3. Charge to the Committee: Dr. Magnuson.
4. Technical and Economic Developments regarding Tetraethyl Lead: Dr. Kirby, Ethyl Corp.
5. Remarks for the DuPont Company: Mr. Sabina and Dr. Zapp.
6. The Medical Problem regarding Tetraethyl Lead: Dr. Kehoe.
7. Committee Discussion.
8. Summary of Action: Dr. Magnuson.

Participants and Observers

Committee:

Dr. James H. Sterner, Medical Director, Eastman Kodak Co.
Dr. Leonard Greenburg, Commissioner, Department of Air Pollution Control, City of New York.
Dr. William P. Yant, Director of Research, Mine Safety Appliances Co.
Dr. Helmuth H. Schrenk, Managing Director, Industrial Hygiene Foundation.

Dr. Stanley H. Osborn, Commissioner of Health, Connecticut Department of Health.
Mr. H. N. Doyle, Assistant Chief, Occupational Health Program, Public Health Service, Reporter.
Dr. H. J. Magnuson, Chief, Occupational Health Program, Public Health Service, Chairman.

Ethyl Corporation:
Dr. Robert A. Kehoe, Medical Consultant.
Dr. George Kirby, Vice President for Research.
Mr. Richard Scales, General Manager, Detroit Research Laboratories.

E. I. duPont de Nemours & Co.:
Mr. J. R. Sabina, Assistant Planning Manager, Division of Petroleum Chemicals.
Dr. John A. Zapp, Director, Haskell Laboratory.

Department of Health, Education, and Welfare:
Mr. Sidney Edelman, Assistant Chief, Public Health Division, Office of the General Counsel.

Public Health Service:
Dr. A. L. Chapman, Chief, Division of Special Health Services.
Dr. D. J. Birmingham, Chief, Clinical Services, Occupational Health Program.
Dr. H. E. Stokinger, Chief, Toxicologic Services, Occupational Health Program.
Dr. W. M. Gafafer, Technical Adviser, Occupational Health Program.
Mr. V. G. MacKenzie, Assistant Chief, Division of Sanitary Engineering Services.
Dr. H. E. Heimann, Assistant Chief, Air Pollution Medical Program.
Mr. J. M. Horstkamp, Chief, Special Services Branch, Bureau of State Services.

Appendix B

Surgeon General's Letter of Transmittal of Report

DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
PUBLIC HEALTH SERVICE

WASHINGTON 25, D.C., *August 21, 1959.*

DEAR _____: I am pleased to transmit the report of an Ad Hoc Committee on Tetraethyl Lead. This Committee was convened at my request to consider the public health hazards which might result if the concentration of tetraethyl lead in motor fuel were to be increased from a maximum of 3 to a maximum of 4 cubic centimeters per gallon of gasoline.

The Committee arrived at two conclusions and made three recommendations. The conclusions are: (1) a change in the maximum concentration of tetraethyl lead in motor fuel from 3.0 to 4.0 cc. would not increase the hazard involved in the manufacture and distribution of leaded gasoline, and (2) available data do not indicate that such a change would significantly increase the hazard to public health from air pollution.

The recommendations are, if the industry does increase the concentration of tetraethyl lead, that:

(1) The industry proceed cautiously with the increase so that the increase in any 1 year in the tetraethyl lead content of U.S. pool gasoline not exceed 0.25 cc. per gallon, and that the concentration of tetraethyl lead in U.S. pool gasoline not be more than 3.2 cc. per gallon at the end of 5 years.

(2) The industry supply annually to the Public Health Service the domestic consumption figures for tetraethyl lead.

(3) The Public Health Service, in collaboration with the manufacturers of tetraethyl lead, the petroleum industry, and other appropriate organizations, conduct studies to provide more definite data on levels and trends of atmospheric lead contamination in selected urban areas and of the body burden of lead of selected population groups. If these studies should bring to light new data of significance, the recommendations of the Committee should be reconsidered.

These recommendations reflect the Committee's expressed concern about the inadequacy of medical data on which to base a conclusive answer to the problem of possible hazard to public health due to lead contamination of the atmosphere.

The Public Health Service transmits these findings to you; however, the report should not be interpreted as a regulation of the Public Health Service or as an agreement between the Public Health Service and industry. Should the decision be made to increase the tetraethyl lead content of motor fuel, I hope that those concerned will seriously consider these recommendations.

We plan to publish the entire committee report which, you will note, contains some technical data supplied by your officials. In addition to such publication we plan to make copies available to the State and Territorial Health Officers and to the Air Pollution Control Association for their comments.

Sincerely yours,

Surgeon General.

Appendix C

Regulations from Public Health Bulletin 163 (1926)

These regulations have been formulated in accordance with the recommendations of the Surgeon General's committee, under whose direction the investigation into the hazards from tetraethyl lead gasoline was conducted. They are proposed for adoption by the several States in order to secure uniformity of control and were the subject of consideration at the meeting of the State and Territorial health authorities with the Surgeon General on May 25, 1926. These regulations are based upon present conditions and knowledge and it is appreciated that changes may be advisable from time to time.

By the term "tetraethyl lead" is meant the chemical substance $Pb(C_2H_5)_4$ of a commercial grade of purity or higher. Ethyl fluid is the concentrated commercial fluid containing tetraethyl lead and other ingredients, which is to be mixed with gasoline to make ethyl gasoline. The manufacture of ethyl fluid consists in adding these other ingredients to tetraethyl lead and is called "blending." The process of adding the ethyl fluid to gasoline to form ethyl gasoline is called "mixing." In the term "ethyl gasoline," as herein used, are included all other motor fluids containing tetraethyl lead.

1. Proposed Regulations for the Manufacture of Tetraethyl Lead and the Blending of the Latter to Make Ethyl Fluid

1. No person shall be employed without adequate instructions as to the nature of the hazard and the precautions to be taken.

2. Each worker shall have a periodical physical examination, which shall consist of such physical and other tests as are indicative of the absorption of tetraethyl lead and shall include, as a minimum, the following items:

(a) Examination of blood for stippling by carefully trained workers, using positive and negative controls, without knowledge of the source of the slides; this examination shall be once a week for the first 3 weeks and bimonthly thereafter.

(b) Semimonthly contact with plant physician for informal statement as to general health.

(c) Bimonthly weight, stripped.

(d) Bimonthly systolic and diastolic blood pressure estimation while sitting.

(e) Bimonthly hemoglobin estimation by Dare's hemoglobinometer.

3. Exact records of examinations shall be kept, and persons showing gradually increasing amounts of stippling, sudden development of stippling, or other marked deviation from normal shall be promptly excluded from tetraethyl lead work, irrespective of whether or not such finding may be indicative of lead poisoning. All parts of the plants where lead in any form is used shall be subject to sanitary measures to prevent collection and dissemination of lead dust.

4. Separate ventilation systems shall be provided for the manufacturing apparatus and for the air of the rooms, the outlets of the latter being located near the floor of each room and all external inlets and outlets being so situated as to avoid dustiness and appreciable contamination of the air around the plant.

5. Daily inspection shall cover efficiency of ventilating systems, all joints, valves, and gaskets of manufacturing apparatus, and adequacy of pressure-hose respirators.

6. All containers of ethyl fluid or tetraethyl lead shall be labeled as to exact content and danger and shall conform to the regulations of the Interstate Commerce Commission. These containers shall be carefully tested for leaks and shall bear a plainly legible label stating that they are to be closed tight immediately when emptied, without cleansing, and sent back to the plant.

7. Kerosene or other material used for cleansing the used containers of ethyl fluid or tetraethyl lead shall be placed in the containers by means of a closed system with air vents to outside air and with adequate ventilation.

8. The filling shall be performed by means of a closed system with air vent from the container to the outside air, and with adequate ventilation.

9. A dye shall be added to ethyl fluid in sufficient amount to give staining qualities to the ethyl gasoline to deter individuals from using it for cleansing or other similar purposes.

10. Reports shall be made monthly to the appropriate officer of the State concerned covering the following points:

Number of workers employed at beginning of month.
Number of workers employed at close of month.
Number of new workers.
Number of workers separated from tetraethyl lead work on account of results of examinations.
Number of definite cases of poisoning.
Condition of cases of poisoning previously reported so far as known.

II. Proposed Regulations for Mixing

1. The maximum content of tetraethyl lead in commercial ethyl gasoline shall be in the proportion of 1 : 1,260 by volume for commercial tetraethyl lead, or 1 : 1,300 for $Pb(C_2H_5)_4$ C.P.

2. Mixing ethyl fluid with gasoline except for certain specific requirements such as research, military and naval use, and airmail service shall be done only at the main distribution centers and in not less than tank-car lots. Adequate provision shall be made at each such center for thorough mechanical distribution of the ethyl fluid throughout the gasoline, and the efficiency of such distribution shall be controlled by the analysis of samples.

3. The locations of these centers and the names of persons engaged in mixing shall be reported to the State department of health of the State of employment.

4. As few persons at each center as practicable shall be employed for this work.

5. No person shall be engaged for mixing until adequately instructed as to the mechanics of mixing, the dangers, and the precautions to be taken.

6. The distributor of ethyl fluid shall provide a special corps of adequately trained instructors and service men.

7. All mixing shall be done with the maximum ventilation practicable under weather conditions existing at the time.

8. Operation of the pumps shall be instantly stopped at the appearance of a leak or other defect, and no attempt shall be made to repair or disconnect the system until a qualified man takes personal charge of it.

9. Floors of all places where any possibility of spilling is present are to be provided with drains and proper facilities for making possible a complete flushing out of all spilled fluid, or in their absence provision shall be made for chemically neutralizing any spillage as it occurs.

10. No bulk mixing station shall be dismantled or disconnected for repairs except by a qualified man.

11. Kerosene or other efficient means of preventing skin absorption of tetraethyl lead and washing facilities shall always be conveniently available.

12. The rules and instructions affecting the employees shall be posted in a conspicuous place where the ethyl fluid is being handled.

13. The distributor of ethyl fluid shall, with the advice of the United States Public Health Service, select a representative group of at least 10 men exposed to the hazard of mixing, representing the entire area of distribution. This group shall serve as an index of the hazard of mixing. Monthly for the first 3 months and every 4 months

thereafter this group of employees shall have an examination, which shall consist of:

(a) Examination of blood for stippling, by carefully trained workers, using positive and negative controls, without knowledge of the source of the slides.
(b) Examination of fecal samples of lead.
(c) Report as to general state of health.
(d) Weight, stripped.
(e) Blood-pressure estimation, diastolic and systolic, when sitting.
(f) Hemoglobin estimation by Dare instrument.

Exact records of these examinations shall be kept.

14. The results of the examination shall be reported promptly to the Surgeon General of the Public Health Service.

III. Proposed Regulations for Distribution of Ethyl Gasoline

1. Each filling station shall have prominently displayed at the pump, or in other conspicuous place, the following warning or one of similar effectiveness: "Ethyl gasoline containing tetraethyl lead, to be used as motor fuel only, and not for cleaning or any other purpose."

2. Suitable leaflets shall be available at all filling stations where ethyl gasoline is sold, for distribution on request. These leaflets shall describe the possible dangers and precautions to be taken in the use of ethyl gasoline.

3. Containers of ethyl gasoline sold to the general public shall be labeled: "Ethyl gasoline containing tetraethyl lead. To be used for motor fuel only, and not for cleaning or any other purpose."

IV. Proposed Regulations for Automobile Garages, Repair Shops, Service Stations and Filling Stations

1. Automobile garages or repair shops holding more than five cars and all inclosed public-service stations and inclosed filling stations for automobiles should be registered with the local health department.

2. In addition to doors and to window ventilation in summer, automobile garages, repair shops, inclosed service stations, and inclosed filling stations should have permanently operative ventilation at least equivalent to that given by an opening or openings at ceiling level free to the outside air, but protected from down drafts, with cross-section of 0.2 percent of the floor space available for cars, together with an opening or openings for inlet near floor level of corresponding effectiveness, both arranged to give positive dilution of exhaust gases as generated. This is to be taken as a minimum, special conditions requiring greater permanent ventilation.

3. Automobile garages, repair shops, inclosed service stations, and inclosed filling stations should be kept reasonably clean and free from

dust by means of flushing out and moist sweeping or vacuum cleaning.

4. Automobile garages, repair shops, service stations, and filling stations should keep conspicuously posted a copy of these regulations with the following statements in large type:

AUTOMOBILE EXHAUST GAS IS DANGEROUS. MOTORS SHOULD NOT RUN LONGER THAN 30 SECONDS UNLESS THE CAR IS IN MOTION OR THE EXHAUST IS DIRECTLY CONNECTED TO THE OUTSIDE AIR. LIQUIDS SOLD AS MOTOR FUEL, EXCEPT ORDINARY GASOLINE, SHOULD BE USED ONLY AS MOTOR FUEL, AND NOT FOR CLEANING OR OTHER PURPOSES. THE FUMES FROM THE EVAPORATION OF EVEN ORDINARY GASOLINE MAY BE INJURIOUS.

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